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Nanoscale magnetic ordering and chemical induced spin freezing in the 2D langasites $Pr_3Ga_5XO_{14}$ (X=Si,Ge,Ti,Sn)

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Abstract – The langasite $Pr_3Ga_5SiO_{14}$ is an example of a frustrated 2D kagomé system with no long ranged magnetic ordering in zero field down to mK temperatures. In applied fields, however, a nanoscale ordering occurs of isolated spin clusters, coexisting with dynamic spins. By chemical doping, one can also induce this spin freezing in zero field by systematically changing the exchange interactions, making this system an important model for tuning spin liquids.

Spins on a kagomé lattice (corner-sharing triangles) remain the canonical example of strongly geometrically frustrated magnetism in two dimensions [1, 2]. These systems have received an increasing amount of attention over the last few decades due to the novel physics that has been observed. Arguably the best studied of these is SCGO ($SrCr_{12-x}Ga_xO_{19}$) [3], which still has a controversial magnetic ground state due to the question of disorder on the chromium sites, and the jarosites, which tend to order at low temperatures [4]. The search is still underway for new 2D spin systems which may be true spin liquids in the limit of zero temperature. The new discovery of the "perfect" kagomé system of quantum, S=1/2, Cu spins ($ZnCu_3(OH)_6Cl_2$) on a lattice of equatorial triangles might be such a candidate [5].

The discovery of spin liquid-like form factors in the new distorted kagomé system $Pr_3Ga_5SiO_{14}$ has renewed interest in the search for highly correlated electron systems with dynamic spins at low temperatures [6]. $Pr_3Ga_5SiO_{14}$ does not have the typical kagomé sublattice, but the topology of the Pr^{3+} spins is equivalent to isolated kagomé planes stacked in the c-direction if one considers the connectivity of nearest neighbor spins. We have recently grown high quality single crystals of this material and have performed extensive characterization measurements to identify the nature of the ground state. The magnetic susceptibility confirms Curie-Weiss behavior down to low temperatures, with no evidence of magnetic long ranged order. The effective magnetic moment of 3.2 μ_B is slightly smaller than the predicted moment of 3.6 μ_B (not uncommon for frustrated systems), and the Weiss temperature is -2.3 K.[7] Specific heat experiments indicate a strong T^2 dependence of the magnetic component, suggestive of 2D dynamics consistent with other kagome systems such as SCGO.[8] With doping, this T^2 component can be diminished in value, either by expanding the lattice (and therefore reducing the exchange interactions), or by the doping of non-magnetic spins on the Pr-sites.

Previous neutron scattering experiments have revealed the presence of short-ranged magnetic correlations building up at low temperatures through a liquid-like magnetic form factor in the elastic channel. Low-energy spin fluctuations were recently measured using the DCS spectrometer at NIST on single crystals. Upon the application of a magnetic field along the c-axis, an unusual partially ordered state develops, and the spin fluctuations are severely suppressed. A Warren-like feature appears as a shoulder upon several Bragg peaks within the HK-plane. This line-shape was originally used to describe two dimensional structural correlations within graphite – disorder exists for the stacked layers along the c-direction and results in anomalous diffuse scattering. The analogy here is that the origin for our diffuse scattering arises from short-ranged ordered Pr^{3+} spins that are disordered from one layer to the next along the c-direction. The identity of this field induced phase is currently unknown – several scenarios have been suggested for this state, from a generalized Kosterlitz-Thouless transition, to a spin nematic phase opening up in applied fields.[7] In this talk, the properties of these new model materials will be discussed as well as future directions for chemical tuning the nanoscale ordering.

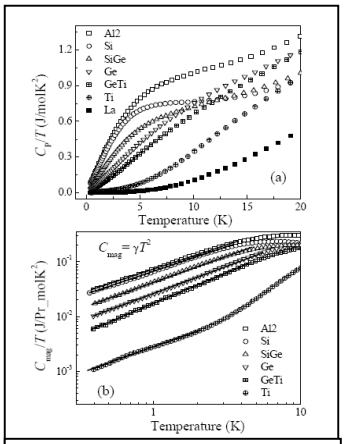

Figure: Suppression of T^2 component of heat capacity through chemical doping. [8]

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